

Evidence of Spin-Injection-Induced Cooper Pair Breaking in Perovskite Ferromagnet-Insulator-Superconductor Heterostructures via Pulsed Current Measurements

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The effect of spin-polarized currents on the critical current densities (J_c) of cuprate superconductors is investigated in perovskite ferromagnet-insulator-superconductor heterostructures with a pulsed current technique. We find that J_c of the superconductor at low temperatures is significantly suppressed as compared with J_c of bare films, and is nearly independent of the temperature (T) and applied magnetic field (H). However, J_c exhibits a slight *increase* with the injection of spin-polarized currents (I_m) from the ferromagnet. In contrast, at higher temperatures near T_c , J_c *decreases* rapidly with increasing T , H , and I_m . These phenomena are attributed to the Cooper pair breaking due to spin-polarized quasiparticle currents transmitted or reflected from the half-metallic ferromagnetic underlayer.

It has been known since the 1960's that the presence of localized magnetic moments in superconductors degrades superconductivity due to the breaking of time-reversal symmetry of the Cooper pairs[1-4], and gives rise to reduction in the superconducting energy gap (Δ) and transition temperature (T_c)[1-3], and to a finite ground-state momentum[4] and quasiparticle density-of-states within the superconducting gap[2,3]. Subsequent experimental studies in the 1970's and 1980's have demonstrated that spin-polarized electrical current from ferromagnets (Fe, Ni, Co) may tunnel through an insulating barrier into a superconductor (Al, Nb)[5-7], and that important physical parameters, such as the degree of spin polarization[5], spin diffusion length (over which the injected electrons lose their spin polarization)[6], and spin lifetime, may be deduced from monitoring the changes in the superconducting properties. These studies have also inspired a new direction of research involving the dynamic process of spin-injection in magnetic materials[7], and has yielded impressive progress in various magnetic devices[7]. Recently, Vas'ko et al.[8] and Dong et al.[9] extended the concept of spin-injection to ferromagnet-insulator-superconductor (F-I-S) heterostructures and demonstrated the suppression of superconducting critical currents in high-temperature superconducting cuprates by injecting electrical currents from the underlying ferromagnetic manganite film. The suppression of superconducting critical currents in those experiments has been attributed to magnetic pair breaking. However, questions remain in that significant current-induced joule heating in those experiments, (typically $10 \sim 100$ mW/cm² in Ref.[8] and up to ~ 1 W/cm² in Ref.[9]), generated by passing a finite current through the resistive ferromagnetic layer, may be the dominant source for the suppression of critical currents.

In order to eliminate the uncertainties surrounding the issue of magnetic pair breaking, we employ in this work a pulsed current technique to ensure that the effect of Joule heating on raising the temperature of the superconductor is reduced to insignificance. Heterostructures with two different compositions have been investigated. They are

$$\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3 - \text{YSZ} - \text{YBa}_2\text{Cu}_3\text{O}_7 \quad (\text{LCMO} - \text{YSZ} - \text{YBCO}),$$

$$\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3 - \text{SrTiO}_3 - \text{YBa}_2\text{Cu}_3\text{O}_7 \quad (\text{LSMO} - \text{STO} - \text{YBCO}),$$

where YSZ is yttria-stabilized-zirconia. These samples are fabricated using pulsed laser deposition on LaAlO_3 (100) (LAO) substrates which are 6 mm \times 6 mm in size. The proce-

dure involves first deposition of the manganite layer on the LAO substrate kept at 700°C under 400 mTorr oxygen, then a thin insulating barrier on top of the masked manganite at 750°C under 200 mTorr oxygen to yield a strip 6 mm long and 1–2 mm wide, and finally YBCO is deposited on the insulating barrier at 800°C and 200 mTorr oxygen. After the deposition, the heterostructures are slowly cooled to room temperature in 500 Torr oxygen. At least two samples of each heterostructure are grown together, so that different characterizations and measurements can be carried out on the same batch of samples. X-ray photoelectron spectroscopy (XPS) has been performed on the surfaces of bilayers to ensure that no visible reaction occurs during the growth process. The thicknesses for both the ferromagnetic and superconducting layers are 100 nm, the YSZ barrier thickness is 1.3 nm, and the STO barrier thickness is 2.0 nm. The morphology of the YBCO surface of the LSMO-STO-YBCO samples is characteristic of c-axis oriented epitaxy, while that of LCMO-YSZ-YBCO samples exhibits some a-axis oriented outgrowths due to the larger YBCO/YSZ lattice mismatch.

For the electrical transport measurements, eight gold contacts, four on YBCO and four on the magnetic layer, are sputtered onto each heterostructure, as illustrated in the top panel of Fig.1. The resistance (R) vs. temperature (T) measurements of each sample are performed with a small direct current ($3\mu\text{A}$), to ensure that no noticeable joule heating occurs in either the YBCO or the magnetic layer. The results on LCMO-YSZ-YBCO and LSMO-STO-YBCO are shown in the middle and bottom panels of Fig.1, respectively. We note that the superconductor and the manganite layer appear to be in good contact, with a junction resistance $< \sim 0.1\Omega$, despite the presence of a thin barrier between them. Consequently, the transport data are displayed in resistance rather than resistivity. However, we note that for both heterostructures, the superconducting layer shows a normal state resistivity ($\rho \approx 60\mu\Omega\text{ cm}$ at the superconducting transition temperature T_c , after accounting for the geometric factors) comparable to that of YBCO single crystals[10] and of YBCO epitaxial films grown directly onto the LAO substrates under similar conditions. In addition, preliminary scanning tunneling spectroscopy studies on the YBCO film of these heterostructures[11] have revealed that the quality of the spectra, in the absence of currents through either YBCO or LCMO (LSMO), are comparable to those of YBCO single

crystals[12] and consistent with *d*-wave symmetry[12,13]. The superconducting transition temperature is $T_c = 86.5$ K for the LCMO-YSZ-YBCO heterostructure, with a resistive transition width $\Delta T_c \approx 1.0$ K, and $T_c(\text{onset}) = 89.0$ K with $\Delta T_c \approx 5.0$ K for LSMO-STO-YBCO. Similar characterizations are also performed on the ferromagnetic layers, as shown in the middle panel of Fig.1 for the LCMO layer at $H = 0, 3, 6$ Tesla, and the bottom panel of Fig. 1 for the LSMO layer at $H = 0$. We find that the Curie temperatures as well as the resistivities of both the LCMO and LSMO layers are comparable to those of the bulk material and epitaxial films on LAO substrates[14], with $T_{\text{Curie}} \approx 260$ K and $\rho(300\text{K}) \approx 15\text{m}\Omega \text{ cm}$ for LCMO and $T_{\text{Curie}} \approx 320$ K and $\rho(300\text{K}) \approx 2\text{m}\Omega \text{ cm}$ for LSMO.

The temperature dependence of the critical current densities (J_c) of YBCO in both LCMO-YSZ-YBCO and LSMO-STO-YBCO, in the absence of any injection current (I_m) from the magnetic layer, are shown in Fig.2(a). Here we have defined the critical current I_c as the current at which the voltage across the length of YBCO is $2 \mu\text{V}$, and J_c is obtained by dividing I_c by the cross sectional area ($2 \text{ mm} \times 100 \text{ nm}$) of YBCO. For comparison, the J_c -vs.- T curve of a typical YBCO/LAO film grown under the same growth condition and with $T_c = 89$ K, is also shown in Fig.2(a). Three important features are noteworthy. First, for both heterostructures, a very large suppression of J_c in the YBCO relative to a typical YBCO/LAO film occurs at low temperatures, and the suppressed J_c is essentially independent of temperature. Second, the J_c of the heterostructure shows a very rapid decrease only at temperatures above $(T/T_c) \approx 0.9 \sim 0.95$, and the magnitude and temperature dependence of $J_c(T/T_c)$ becomes comparable to those of the YBCO/LAO film. Third, the low-temperature J_c values are independent of the magnetic field (H), while the high-temperature J_c values show a significant suppression due to the presence of a magnetic field parallel to the YBCO layer, as illustrated in Fig.2(b).

To investigate the effect of injection current from the magnetic layer, and to prevent distortion of results due to heating of YBCO from power dissipation in the magnetic layer, we synchronize the periods of two pulse generators which supply pulsed currents to the YBCO and LCMO (LSMO) layers, and choose a pulse width which yields a negligible temperature rise ($< 10 \text{ mK}$) in the YBCO, as determined by monitoring the changes in the resistance of the ferromagnetic layer during the measurements. For each value of the

current through the ferromagnetic layer, I_m , the corresponding critical current I_c through YBCO is obtained from the relation $I_c = (I_c^+ - I_c^-)/2$, where I_c^+ and I_c^- are defined as the currents at which the voltages along the YBCO film are $+2\mu\text{V}$ and $-2\mu\text{V}$, respectively. As illustrated in Fig.3 for data taken on the LCMO-YSZ-YBCO heterostructure at a nominal temperature of $T = 84.2\text{K}$ indicated by a carbon-glass temperature sensor adjacent to the sample, we find that continuous direct currents through LCMO, with a maximum power dissipation $\sim 10\text{ mW/cm}^2$ at $I_m = 15\text{ mA}$, yield a large suppression of the critical current I_c . The suppression of I_c is significantly reduced under a pulsed current with a width $200\text{ }\mu\text{s}$ and a period 200 ms , which correspond to a power dissipation $< 400\text{ }\mu\text{W/cm}^2$ for the injection current $I_m = 100\text{ mA}$, and the corresponding temperature rise as monitored by our *in-situ* thermometry, the resistance of the LCMO, is less than 10 mK . Hence, our subsequent studies of the J_c -vs.- I_m isotherms are measured using pulsed currents with a width-to-period ratio of 10^{-3} . It is important to note that a current-induced power dissipation of $\sim 10\text{ mW/cm}^2$, which is comparable to that in Ref.[8] and is two orders of magnitude smaller than that of $\sim 1\text{ W/cm}^2$ in Ref.[9], has been shown in Fig.3 as already too large to yield conclusive information for the dependence of the critical current on spin injection.

The J_c -vs.- I_m measurements indicate that J_c is nearly independent of I_m at low temperatures, whereas a significant suppression of J_c is observed with increasing I_m at temperatures closer to T_c , as shown in Fig. 4 for the LSMO-STO-YBCO sample, and in the inset of Fig.4 for the LCMO-YSZ-YBCO sample. Furthermore, in the presence of a finite dc magnetic field parallel to the layers, J_c at low temperatures still remains invariant under a finite I_m . In addition, a general trend of a slight increase in J_c with the initial increase of I_m is always present before a significant decrease at sufficiently large I_m and at high temperatures (see Fig.4).

To better understand the physical implications of the above results, we first consider the pronounced suppression of the low-temperature J_c in YBCO as compared with J_c of bare YBCO films (see Fig.2(a)). Noting that *both the normal-state resistivity of YBCO in the heterostructure and its superconducting gap are comparable to those of the single crystalline samples*[10-12], we conclude that the quality of the samples is high, and we

shall suggest in the following that observed suppression of low-temperature J_c in the F-I-S heterostructures is a manifestation of Cooper pair breaking due to the reflected spin-polarized quasiparticle currents from the interface with a half-metallic ferromagnet.

It is reasonable to assume that an applied current I_s in YBCO allows a finite leakage current through the interfaces and into the ferromagnetic half metal, which can be understood as follows. Measurements of J_c are performed using a finite voltage criterion, ($V = 2 \mu\text{V}$ in our case). This finite voltage corresponds to a very small but non-zero resistance in the YBCO film for $I_s \approx I_c$, and therefore a small fraction of I_s can flow through the interface into the magnetic underlayer. Since the ferromagnetic manganites are half metals with complete spin polarization[14-17], only quasiparticles with spins parallel to those of the carriers in the manganite are transmitted across the interface, leaving a spin-polarized quasiparticle current in YBCO and resulting in substantial pair breaking. The pair breaking induced by the spin-polarized current further increases the resistance of YBCO and results in larger leakage current, and therefore more pair breaking. This avalanche-like process is limited, however, by the fact that enhanced pair breaking and consequent increased population of quasiparticles have an effect on suppressing further pair breaking[4], because quasiparticles are Fermions and obey the exclusion principle. Hence, a small but non-zero values of J_c is measured in the F-I-S heterostructures. Similarly, the absence of significant dependence of the low-temperature J_c on either H or I_m may be understood in the same context. That is, if the reflected spin-polarized currents already achieve the maximum pair-breaking effects, there will be little effect on J_c by applying more spin-polarized currents I_m or an external magnetic field H . On the other hand, at temperatures near T_c , large thermal fluctuations of the superconducting order parameter is likely to reduce the degree of spin polarization substantially in the reflected quasiparticle current. Hence, J_c in the heterostructure becomes comparable to that of YBCO/LAO films if $I_m = 0$ and $H = 0$, as shown in Fig.2(a). With the application of either I_m or H , the degree of spin polarization (and therefore the effects of magnetic pair breaking) is enhanced against thermal fluctuations of the superconducting order parameter. Consequently, J_c becomes strongly affected by I_m and H at temperatures near T_c .

More specifically, we may argue that the critical current I_c is uniquely determined

by T , H , and the total spin-polarized quasiparticle current I_m^{tot} in the superconductor. Furthermore, I_m^{tot} consists of contributions from both the external spin injection current (I_m) and the transport current (I_s) in the superconductor, and may be given by:

$$I_m^{tot}(T, H, V) = \eta_t(T, H, V)I_m + \eta_r(T, H, V)I_s, \quad (1)$$

where V is the bias voltage between the ferromagnet and the superconductor, η_t denotes the fraction of spin-polarized current transmitted from the ferromagnetic layer to the superconductor, and η_r is the fraction of spin-polarized quasiparticle currents reflected from the ferromagnetic interface. We expect that both η_t and η_r are dependent on the interface properties and the density of states of the ferromagnetic and superconducting layers. In addition, the degree of spin polarization in the ferromagnet, as well as the spin diffusion length and the spin life time in the superconductor, are all important factors in determining the coefficients η_t and η_r . It is therefore reasonable to assume that η_r increases with decreasing temperature and increases with increasing magnetic field, because a higher degree of spin-polarization is expected at low T and high H . On the other hand, the temperature and magnetic field dependence of η_t is more complex. For instance, a sub-gap spin-polarized electron injected from the ferromagnet can either be normally reflected back or Andreev reflected[18] as a hole with an opposite spin. The latter possibility cannot be realized in the case of a half-metallic ferromagnet that exhibits 100 % spin polarization. It should also be noted that the Cooper pairs transmitted into the superconductor as a result of Andreev reflection do not contribute to the total spin polarized current I_m^{tot} , because each Cooper pair forms a singlet state with two opposite spins. Therefore only high-energy (*i.e.*, with energies higher than Δ) spin polarized electrons contribute to the first term $\eta_t I_m$ of Eq.(1). However, we remark that the $d_{x^2-y^2}$ pairing symmetry in YBCO[12,17] implies the existence of infinitesimally small superconducting gaps near the $\{110\}$ direction of momentum space. Consequently, the transmission of the spin polarized current from the ferromagnet to the superconductor is likely to be more effective in the case d -wave superconducting cuprates than in the case of conventional s -wave superconductors.

Following the conjecture given by Eq.(1), a finite transport current I_s in the superconductor can result in a finite spin polarized current $\eta_r I_s$ even if $I_m = 0$. On the other hand,

as η_r decreases with temperature, the pair breaking mechanism at high temperatures is primarily due to thermal effects rather than spin polarized currents if $I_m = 0$. Hence, in the absence of I_m , J_c values for YBCO in the F-I-S heterostructures become comparable to those of the bare YBCO films near T_c , as shown in Fig.2. In other words, $I_m^{tot} \rightarrow 0$ near T_c unless $I_m \neq 0$ or $H \neq 0$. Consequently, the presence of an external I_m or an applied magnetic field has a much more significant effect on suppressing I_c if $T \rightarrow T_c^-$.

Finally, we comment on the anomalous increase in I_c with the initial increase of I_m . We note that an excess voltage drop across the ferromagnet layer, as the result of a finite I_m and finite resistance in the ferromagnet, effectively offsets the chemical potential of the ferromagnet relative to the superconductor, so that $V \neq 0$. This offset may impede quasiparticle transport from the superconductor to the ferromagnet, thereby yielding a slight decrease in η_r . Hence, I_m^{tot} is reduced, giving rise to a small increase in I_c . However, the slight decrease in the component $(\eta_r I_s)$ is eventually overwhelmed by the other increasing component $(\eta_t I_m)$, so that I_c decreases rapidly with increasing I_m , as shown in Fig. 4.

In summary, we have demonstrated experimental evidence for spin-injection-induced Cooper pair breaking in perovskite F-I-S heterostructures by employing pulsed current technique to efficiently minimize Joule heating. In the absence of an external injection current from the ferromagnetic layer to the superconductor, a large suppression of the low-temperature critical current density J_c is observed in the heterostructure. We attribute the suppression of the low-temperature J_c to Cooper pair breaking incurred by spin-polarized quasiparticle currents that are reflected from the interface of YBCO with the underlying half-metallic ferromagnet. This suppressed low-temperature J_c is found to be insensitive to the applied magnetic field H and temperature T . However, the low-temperature J_c appears to increase slightly with the increasing spin-injection current I_m . In contrast, J_c at high temperatures shows a strong dependence on both I_m and H , and a slight increase in J_c is observed before rapid drop-offs with increasing I_m . Our findings are qualitatively consistent with the scenario of Cooper pair breaking under spin-polarized currents, although thorough quantitative understanding of these results still awaits further experimental and theoretical investigation.

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Figure Captions

Fig.1 (a) Schematics of the ferromagnet-insulator-superconductor heterostructures and the electrical contact configurations. The dark strips represent the gold electrical contacts on the ferromagnetic and the superconducting layers. (b) The resistance (R) vs. temperature (T) curves of the YBCO (left axis) and those of the LCMO (right axis) for $H = 0, 3, 6$ T in the LCMO-YSZ-YBCO heterostructure. (c) The R -vs.- T curves of the YBCO (left axis) and LSMO (right axis) in the LSMO-STO-YBCO heterostructure.

Fig.2 (a) Comparison of the zero-field critical current density (J_c) of YBCO, LCMO-YSZ-YBCO and LSMO-STO-YBCO films as a function of the reduced temperature (T/T_c). (b) $J_c(T)$ for LSMO-STO-YBCO at $H = 0$ and 3 T.

Fig.3 Significant joule heating due to injection current (I_m) on the measured critical current (I_c) is illustrated by comparing the I_c -vs.- I_m data for continuous dc current and for pulsed currents. The nominal temperature of the measurements indicated by a carbon-glass temperature sensor is $T = 84.2$ K, and the sample is LCMO-YSZ-YBCO.

Fig.4 Effects of spin injection on the J_c of LSMO-STO-YBCO are illustrated by comparing the $J_c(T)/J_c^0(T)$ -vs.- I_m curves at various temperatures. Here $J_c^0(T)$ denotes the critical current density under no injection current. The inset shows the corresponding results for the LCMO-YSZ-YBCO heterostructure.







